

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Richard HERTZOG et al.

Application No: 08/601,879

Group Art Unit: 1205

Filed: February 15, 1996

Examiner: J. Reamer

For: DECOMPOSITION OF CUMENE OXIDATION PRODUCT

DECLARATION UNDER 37 CFR 1.132

Assistant Commissioner of Patents  
Washington, D.C. 20231

Sir:

I, Stylianos Sifniades, declare as follows:

(1) I am a named coinventor of the above-identified application.

(2) In 1957, I received a Diploma of Chemistry from the University of Athens, Greece. In 1962, I received an M.S. in Physical Chemistry from the University of British Columbia, Canada. In 1965, I received a Ph.D. in Physical Chemistry from the University of British Columbia, Canada.

(3) Since 1965, I have been employed by AlliedSignal (formerly Allied Chemical). In 1965, I joined Allied Chemical (now AlliedSignal) as a Sr. Research Chemist. From 1968 to 1976, I held the position of Research Group Leader. From 1976 to 1980, I held the position of Research Supervisor. Since 1980 (to present), I have held the position of Research Fellow.

(4) Fields in which I have been involved include computer simulation of chemical and physical processes, studies of chemical kinetics and chemical process development (e.g., phenol, acetone,

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alpha-methylstyrene, caprolactam and nylon 6). I have been actively involved in the development, simulation and optimization of the cumene to phenol process and downstream applications since 1979. My work was instrumental in the optimization and expansion of AlliedSignal's phenol plant at Frankford, PA (current capacity ca 900Mlb/yr) and in the licensing of the phenol process to others. I have made numerous technical presentations on this subject, and I am recognized as the foremost expert in this field within AlliedSignal and its licensees (e.g., Rhone-Poulenc of France, Kumho of Korea and Ertisa of Spain).

(5) In 1986, I received an AlliedSignal Technical Achievement Award - Improved Phenol Process. In 1995, I received an AlliedSignal Special Achievement Award - Carpet Fiber Recycling. I am a member of the American Chemical Society.

(6) I am an author or coauthor of 19 technical papers, and I am a named inventor or coinventor in 35 U.S. patents. The following are particularly relevant to the field of phenol chemistry:

- (a) Sifniades, A.A. Tunick and F.W. Koff "Decomposition of Cumene Oxidation Product" US Patent 4,358,618;
- (b) Sifniades, Chapter "Acetone" in Ullmann's Encyclopedia of Industrial Chemistry, 5th Edition, Vol. A1, pp. 79-96, VCH Publishers (1985); and
- (c) R. Pujado and S. Sifniades, Chapter "Phenol" in

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Encyclopedia of Chemical Processing and Design, Vol. 35, pp 372-391, Marcel Dekker Inc (1990).

(6) I am listed in American Men and Women of Science - Physical & Biological Sciences, 16th Edition (1986) Vol. VI.

(7) It is not obvious to arrive at an embodiment of the present invention by combining the disclosures of the Sifniades et al U.S. Patent No. 4,358,618<sup>1</sup> and the Barilli British Patent No. 1,202,687. The respective processes described in these patents involve a host of conflicting features. It would not be obvious to pick and choose from among these diverse features, while selecting certain features and ignoring other features without any particular guidance or motivation to make such choices.

(8) Features of the Barilli patent include, among others, the following:

(a) the entire reaction is conducted in a single vessel at a temperature which never exceeds 70°C;

(b) a very large amount of acid catalyst is employed in the reaction (e.g. 1,500 ppm of sulfuric acid is used in the reaction mixtures of the Examples); and

(c) acetone in excess of that produced in the decomposition reaction is introduced into the reactor.

(9) Features of the Sifniades et al patent include, among

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<sup>1</sup> I am also a coinventor of this U.S. Patent No. 4,358,618.

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others, the following:

- (a) the entire reaction is conducted in multiple reaction vessels at a temperatures which exceed 120°C during the later stages of the reaction;
- (b) a very small amount of acid catalyst is employed in the reaction (e.g. 50 ppm of sulfuric acid is used in the reaction mixtures of certain Examples); and
- (c) no excess acetone in addition to that produced in the decomposition reaction is introduced into the reactor.

(10) There is no motivation in the respective disclosures to combine and/or substitute features from the Sifniades et al patent with features from Barilli patent. The Barilli patent suggests that, when the reaction mixture contains high concentrations of sulfuric acid catalyst and the decomposition is carried out to completion in a single reaction vessel, to minimize by-product formation, it is necessary to maintain the maximum temperature of the reaction below 70°C and to feed excess acetone to the reaction mixture. The purpose of these controls is said to be for the prevention of sulfuric acid from catalyzing condensing and dehydrating reactions. At page 2, lines 18-20, it is specifically warned that the disadvantageous effects of sulfuric acid are promoted by temperatures in excess of 70°C.

(11) Unwanted by-product formation is prevented in an entirely different manner in the process disclosed in the Sifniades

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et al patent. For example, the deleterious effects of the catalyst are minimized by employing far less of the catalyst and by using a water moderator. Under these conditions, temperatures in excess of 120°C can be used at the end of the reaction to obtain greater amounts of alpha-methylstyrene (AMS).

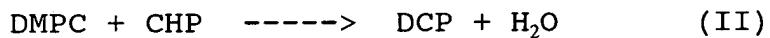
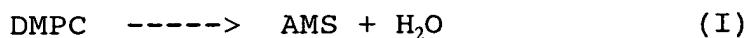
(12) In accordance with the present invention, it was surprisingly discovered that the yields of AMS can be even further enhanced by employing excess acetone in the reaction mixture. Additionally, it was discovered that by employing excess acetone in the reaction mixture enhanced yield of AMS can be obtained at relatively low concentrations of residual CHP in step (a) of the Sifniades et al patent, thus, increasing the stability of the process (this point is further elaborated upon hereinafter in paragraph 22). These results are nowhere suggested in the disclosures of the Sifniades et al patent and the Barilli patent, whether these disclosures are viewed singly or in combination.

(13) The disclosure of the Sifniades et al patent is silent regarding the results achieved by the use of excess acetone. The results achieved by the use of excess acetone in the Barilli patent pertain to an entirely different set of conditions than those employed in the Sifniades et al patent. Therefore, it is uncertain that the same results would be achieved. Furthermore, even assuming, *arguendo*, that the same results would be achieved such results would be contrary to the results desired in the Sifniades

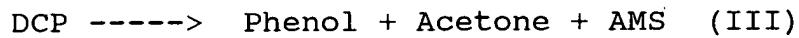
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et al patent.

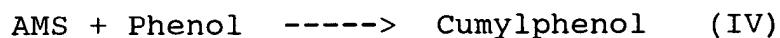
(14) The Barilli patent teaches that condensation and dehydration reactions are to be suppressed. However, AMS is formed by such condensation and dehydration reactions. These reactions include the following:



Accordingly, by restricting the reaction (I), the formation of AMS is directly restricted, and by restricting the reaction (II) the formation of AMS is further restricted by preventing the initial step of the following sequence of reactions:



By restricting the formation of AMS, the formation of the unwanted by-products, cumylphenol and AMS dimers, is also prevented according to the following reactions:



Accordingly, the Barilli patent teaches that the root cause of by-product formation should be completely cut off at the source by preventing the formation of AMS via condensation and dehydration reactions.

(15) If anything, then, adding the teaching of the Barilli

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patent to that of the Sifniades et al patent would suggest that the addition of excess acetone to the reaction mixture of the Sifniades et al patent would suppress the formation of AMS. Surprisingly, however, it has been discovered that such acetone addition actually enhances the formation of AMS.

(16) Evidence of this surprising AMS formation is provided in the Examples of the present application. The results are summarized in Figure 2, which provides graphs showing the results of excess acetone addition.

(17) In a decision (Paper No. 16), mailed March 18, 1992, by the Board of Patent Appeals and Interferences in the grandparent application (SN 08/297,333), these results are discussed. The following statement is first noted:

"First, while it appears that the data in question is intended to reflect the difference between a process of the type taught by Sifniades with versus without the addition of acetone, the difference established by the data are not described in the specification with sufficient detail to determine whether they are statistically significant, unexpected and of practical significance."

However, I respectfully disagree with this statement.

(18) It is apparent on the face of Figure 2 that there is a statistical difference between the results achieved with acetone recycle and without acetone recycle. As pointed out hereinabove,

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e.g., in paragraphs 9-11, these results are unexpected. Clearly these results are of practical significance. AMS is a valuable composition of matter, and as suggested in the Abstract of the Sifniades et al patent, it is desired to maximize its production.

(19) The following statement in said decision is also noted: "Secondly, it appears to us that any improved product yield reflected in this data would have been expected rather than unexpected. That is, since the multistage process of Sifniades and the acetone addition of Barilli are disclosed as individually resulting in improved yields, the combination of Sifniades' multistage process with Barilli's acetone addition would have been expected to result in an even greater yield improvement."

However, I respectfully disagree with this statement.

(20) As pointed out hereinabove, e.g., in paragraphs 9-11, the results, in terms of enhanced AMS yields, are unexpected. If anything, the Barilli patent would suggest that excess acetone addition would result in decreased AMS yields.

(21) The following statement in said decision is also noted: "Thirdly, even if the appellants' data were assumed to evince unexpected results, this evidence is plainly not commensurate in scope with the claims to which it pertains (cf., the 40 percent and 60 percent concentrations of this data and the 10 percent to 100 per cent acetone concentrations of the appealed

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claims) and therefore is inadequate to rebut the examiner's prima facie case of obviousness."

However, I respectfully disagree with this statement.

(21) It is appropriate to extrapolate the results of the present Examples. Clearly the advantageous results do not begin at 40 percent and end at 60 percent. The claimed range of 10 to 100 percent is reasonably supported by these data. The claimed range of 10 to 100 is further supported by data given in the Zakoshansky U.S. Patent No. 5,254,751, especially in Table 3 on column 9.

(22) A further elaboration on the nonobvious effect of acetone addition on reactor stability, as referred to above in paragraph 12, is as follows. The process taught by the Sifniades et al patent relies to a great extent on the above-identified reactions (II) and (III). Reaction (II) is first order with respect to CHP, therefore, the higher the concentration of residual CHP in step (a) of the process, the larger the extent to which DMPC is converted to DCP and the higher the yield of AMS obtained in step (c) of the process. Yields of AMS over 80 mol% of DMPC can be achieved by allowing residual CHP concentrations of 3 wt% or higher in the reactor of step (a). However, high residual CHP concentrations may result in an unstable reactor unless large and expensive cooling systems and elaborate feedback loops are employed. Each weight percent of CHP that decomposes adiabatically in the reaction mixture causes a temperature rise of ca 7°C.

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Additionally, the rate of CHP decomposition roughly doubles for every 6°C of temperature rise. Thus, the potential exists for a runaway reaction. The reactor temperature is controlled by balancing the rate of heat removal with the rate of heat formation. Generally, the higher the potential for exotherm in a commercial reactor, the larger must be the capacity of the cooling system that is used to maintain stability. At CHP concentrations of about 3 wt%, a large and very expensive cooling system would be required to compensate for the large release of thermal energy accompanying CHP decomposition that may be triggered by a hot spot, a local surge in catalyst concentration or other events. At still higher CHP concentrations it may be impossible to control the reactor even with an expensive cooling system. It is, therefore, desirable to maintain a low level of residual CHP in the reactor while achieving a high AMS yield. According to the teachings of the Sifniades et al patent (sentence bridging columns 3 and 4) the level of residual CHP can be adjusted by employing the appropriate combination of temperature, residence time, catalyst concentration and water concentration. There is no mention of acetone addition. We have surprisingly found that if acetone is added to the reactor of step (a) of said patent, reactions (II) and (III) are promoted at generally lower levels of residual CHP than in the absence of added acetone. Thus, high yields of AMS are obtained in step (c) without jeopardizing the stability of the reactor of step (a). There is no

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teaching in the Barilli patent that acetone should be added to a multistage reactor.

(23) The irrelevance of the Barilli patent to the process of the present application is further indicated by the fact that the highest yield of phenol shown in the Examples of the Barilli patent is 95.8%. The yield of phenol achieved in the process of the Sifniades et al patent is typically over 98.5%. It is not obvious that adding acetone to step (a) of the process of the Sifniades et al patent will improve the phenol yield beyond the level taught in the Barilli patent.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

  
Stylianos Sifniades

Date: June 7, 1996